

**Answer**

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#### 4. Date of Notification

International Searching Authority Opinion: December 7, 2004

#### 5. Contents of Answer

On the basis of the comment (in the written opinion issued by the PCT International Searching Authority, dispatched on December 7, 2004) that the inventions according to Claims 1 to 6 and 10, among Claims 1 to 11, lack in novelty and inventive step because of the following documents cited in the international search report, the applicant has amended Claims 1 and 3 and expresses an opinion as follows.

Document 1: JP-A-2002-198050A

Document 2: JP-A-2003-34534A

##### (1) Regarding the Invention according to Claim 1 (amended)

##### (1-1) Contents of Amendment

Claim 1 after amendment reads as follows. Note that the underline indicates the amended part.

"1. (Amended) A cathode material for a secondary battery containing a cathode active material represented by a general formula  $\text{Li}_n\text{FePO}_4$  (wherein n represents a number from 0 to 1) as a primary component, one or more metal elements selected from the group consisting of metal elements belonging to Group 4, 5, 6, 11, 12, 13 and 14 of the Periodic Table, the total content of which is in the range of 0.1 to 5 mol%, in terms of element ratio, based on iron in the cathode active material, and a halogen

element in an amount of 0.1 mol% or more based on P.”

(1-2) Basis of Amendment

The basis of the amendment is Claim 3 before amendment.

(1-3) Characteristics of the Invention

The invention according to Claim 1 is characterized in that:

<1> a cathode material for a secondary battery containing a cathode active material represented by a general formula  $\text{Li}_n\text{FePO}_4$  (wherein n represents a number from 0 to 1) as a primary component is provided;

<2> the cathode material contains one or more metal elements exclusively selected from the group consisting of metal elements belonging to Group 4, 5, 6, 11, 12, 13, and 14 of the Periodic Table;

<3> the total content of the metal elements is exclusively in the range of 0.1 to 5 mol%, in terms of element ratio, based on iron in the cathode active material; and

<4> the cathode material contains a halogen element in an amount of 0.1 mol% or more based on P.

In other words, the invention of Claim 1 is characterized in that: the cathode material does not contain any metal element belonging to Group 7 to 10, such as Mn, Co, Ni, etc., of the Periodic Table but contains exclusively the above-mentioned metal elements (Group 4 to 6 and 11 to 14); and the cathode material contains a halogen element in an amount of 0.1 mol% or more based on P.

In this way, by containing exclusively specific metal elements and containing a halogen element in a specific amount, the cathode material can exhibit remarkably improved cycle characteristics in discharge capacity of resulting secondary batteries, as described in relation to Examples 1 to 10 and FIGs. 5 to 14 corresponding thereto.

The cathode materials of Comparative Examples 1 to 5, of which data are shown FIGs. 5 to 14, do contain the above-mentioned metal elements (Group 4 to 6 and 11 to 14), but do not themselves contain halogen, as can be understood from the fact that their ingredients do not contain halogen. The cycle characteristics of these cathode materials containing no halogen deteriorate with the progress of the cycles, similarly to those of the cathode materials containing no above-mentioned metal elements.

Again, the cathode materials of Examples 1 to 10 of the present invention which contain halogen exhibit remarkably improved cycle characteristics, as clearly shown in Table 4, compared to those of cathode materials containing no halogen.

[Reference Figure 1]

Reference Figure 1 adds to the basis of the cathode material of the present

invention containing no metal elements belonging to Group 7 to 10 of the Periodic Table, such as Mn, Co, Ni, etc., but containing exclusively the above-mentioned metal elements (Group 4 to 6 and 11 to 14).

The cycle characteristics of the cathode materials containing Mn, Co, Ni or Mg deteriorate significantly, compared to those of the cathode materials containing the metal elements according to the present invention (Cu and Zn in this figure). Incidentally, each of the cathode materials contains chlorine, as halogen, in an amount of 0.8 to 1.1 mol% based on P.

In this way, Reference Figure 1 has successfully explained the significance of the cathode material containing no metal elements belonging to Group 7 to 10 of the Periodic Table, such as Mn, Co, Ni, etc., but containing exclusively the above-mentioned metal elements (Group 4 to 6 and 11 to 14).

[Reference Figure 2]

Reference Figure 2 adds to the basis of the total content of the metal elements being exclusively in the range of 0.1 to 5 mol%, in terms of element ratio, based on iron in the cathode active material. Compositing the metal elements improves the battery performance such as conductivity and polarization of the cathode material itself. However, since the metal elements are substituted for Li or Fe which constitutes an active material in the resulting battery, the capacity which depends on the active material deteriorates.

The cathode material represented by the uppermost cycle characteristics line in Reference Figure 2 contains metal Cu in a total amount of 1 mol%, substituted for 0.5 mol% of Li and Fe, respectively. The other cycle characteristics lines represent cathode materials containing Cu in five times that amount, or in an amount of 5 mol%. It is shown that the initial discharge capacity deteriorates with a larger amount of Cu. For this reason, 5 mol% would be the upper limit. Incidentally, Reference Figures 1 and 2 also show the content of halogen (chlorine).

#### (1-4) Comparison with Document 1

Document 1 discloses in its Paragraph [0016] that the cathode material contains at least one metal element other than Fe, Li or Al, among which Co, Mn and Ni are preferable. Hence, the cathode material of Document 1 can extensively contain almost any metal element. Document 1 also discloses that the content of the metal element is  $0 \leq y \leq 0.5$ , which even permits a content about one order larger than that in the present invention.

In other words, Document 1 treats the metal elements specified in the present invention (Group 4 to 6 and 11 to 14) and those not specified in the present invention

on the basis of the fact known from Reference Figure 1, or metal elements belonging to Group 7 to 10, such as Mn, Co, Ni, etc. in the same way as each other as metal elements to be contained in the cathode material.

In addition, while the content of the metal elements is exclusively in the range of 0.1 to 5 mol%, in terms of element ratio, based on iron in the cathode active material on the basis of the fact known from Reference Figure 2 in the present invention, Document 1 permits a wider range from below the lower limit to above the higher limit.

Moreover, Document 1 discloses that the cathode material contains halogen. It discloses that the halogen element is substituted for oxygen atoms of  $\text{PO}_4$  constituting the cathode active material (Paragraphs [0020], [0055], etc.) and that this substitution distorts the crystal framework and hence improves the charge/discharge characteristics at larger currents (Paragraph [0008]), but discloses no further than that.

While Document 1 does disclose that the substitution distorts the crystal framework, it does not explain the basis of how the distortion actually occurs. Furthermore, as can be seen in the specific examples of composition in Table 1, cathode materials containing halogen substituted for oxygen atoms of  $\text{PO}_4$  are far from common compounds and could not be manufactured (materialized) in common manufacturing methods such as described in Paragraph [0022]. Thus, Document 1 can be safely concluded as having little basis of its content.

#### (1-5) Patentability of the Invention

Document 1, when understood not superficially but independently and thoroughly, can be reasonably construed as not describing or suggesting the teaching of the present invention, more specifically the teaching of the invention according to Claim 1, that the cathode material contains:

exclusively specific metal elements (not including Mn, Co, Ni, etc.); and  
a halogen element in a specific amount,  
thereby exhibiting remarkably improved cycle characteristics in discharge capacity of resulting secondary batteries (FIGs. 5 to 14).

As has been described in the foregoing, the invention according to Claim 1 is different from the invention described in Document 1 and cannot be easily made based thereon.

#### (2) Regarding the Invention according to Claim 2

In Claim 2, the metal elements of Claim 1 are limited to specific metal elements

described in Examples. The invention of Claim 2 discloses that the cycle characteristics in discharge capacity of resulting secondary batteries can be remarkably improved by such metal elements and the halogen (FIGs. 5 to 14). Document 1 does not describe or suggest the specific constitutions or effects peculiar thereto.

(3) Regarding the Invention according to Claim 3 (amended)

Claim 3 after amendment reads as follows. Note that the underline indicates the amended part.

“3. (Amended) The cathode material for a secondary battery according to Claim 1 or 2, wherein the halogen element is in an amount of 0.1 mol% or more based on P and in a molar concentration of up to twice that of the metal element or elements.”

The basis of the amendment is Paragraph [0193].

The invention of Claim 3 specifies the quantitative range of halogen in the cathode material.

Document 1 does not describe such a quantitative range of halogen.

(4) Regarding the Invention according to Claim 4

The invention of Claim 4 discloses a cathode material for a secondary battery synthesized so as to contain a cathode active material represented by a general formula  $\text{Li}_n\text{FePO}_4$  (wherein n represents a number from 0 to 1) as a primary component and one or more metal elements by mixing a halide or halides of the metal element or elements and the ingredients of the cathode active material and calcining the mixture.

The invention of Claim 4 is characterized in that a halide or halides of the metal element or elements are used as the ingredients. In this way, a cathode material can be easily produced which contains the metal element or elements and also the halogen element or elements (as residues). The thus produced cathode material for a secondary battery is expected to yield improved cycle characteristics.

Document 1 does not describe the production method in detail, and does not describe the use of a halide or halides of a metal element or elements. Its Paragraph [0026] contains a description reading “element, organic compound, hydroxide, oxide, carbonate, nitrate, complex, etc.,” but no more than that. Needless to say, Document 1 does not disclose or suggest that the use of such a halide or halides as the ingredients can result in the existence of a metal element or elements and a halide or halides (as residuals) and in improved cycle characteristics.

(5) Regarding the Invention according to Claim 5

It is considered that the invention of Claim 5, of which inventive step has been denied after additional consideration of Document 2, has patentability since the inventions according to Claims 1 to 4 have patentability as described above.

(6) Regarding the Invention according to Claim 6

It is considered that the invention of Claim 6 has patentability for the same reason as the invention of Claim 4.

(7) The patentability of Claims 7 to 9 and 11 has not been denied.

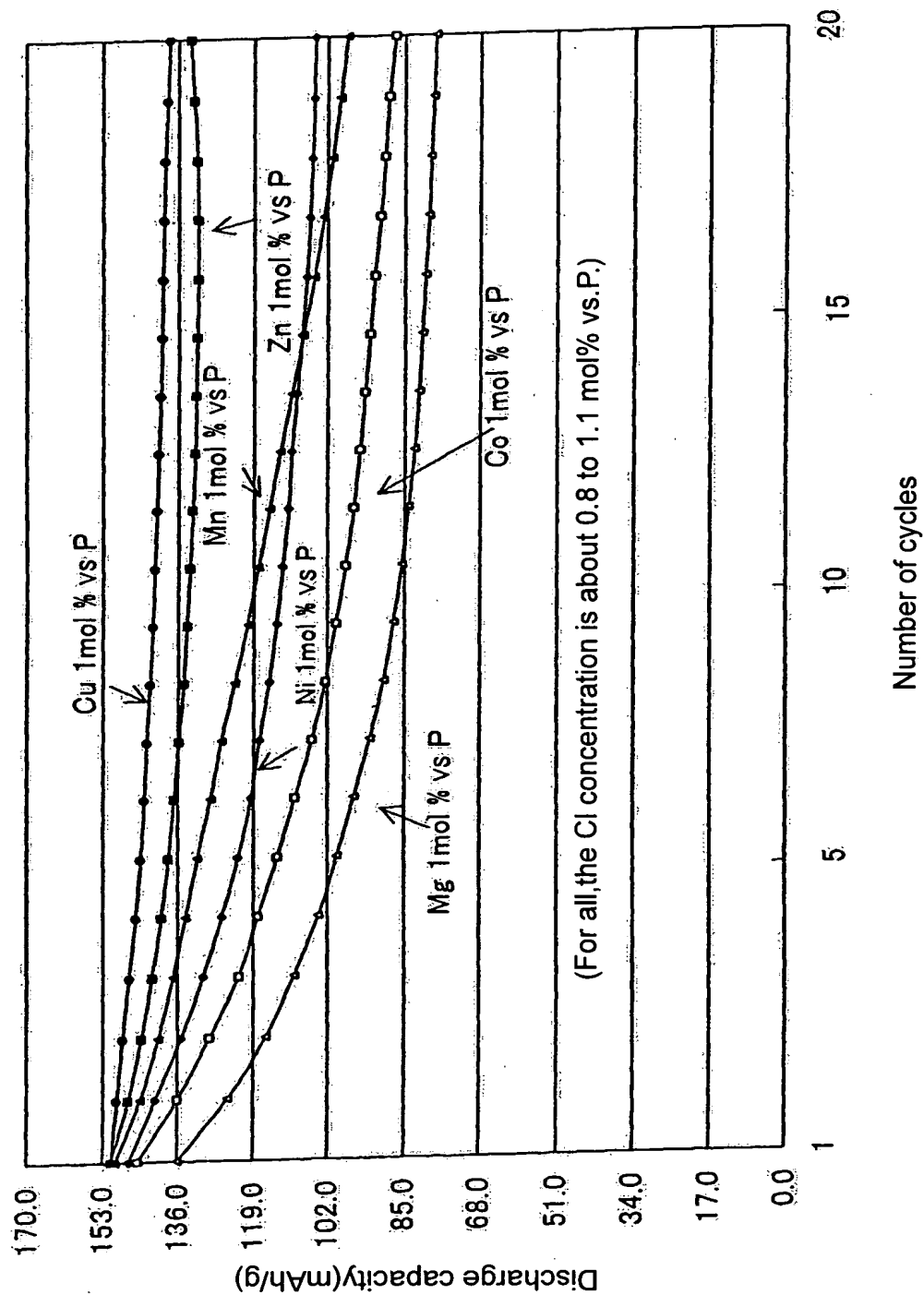
(8) It is considered that the invention of Claim 10 has patentability since the inventions according to Claims 1 to 5 have patentability as described above.

6. Attached Documents

(1) Reference Figure 1

(2) Reference Figure 2

[Reference Figure 1]





[Reference Figure 2]

